

Superconducting order parameter in heavily overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$: a global quantitative analysis

R. H. He^{‡,1} D. L. Feng,^{1,*} H. Eisaki,² J.-i. Shimoyama,³ K. Kishio,³ and G. D. Gu⁴

¹*Physics Department, Applied Surface Physics State Key Laboratory,
and Synchrotron Radiation Research Center,
Fudan University, Shanghai 200433, China*

²*Nano-electronic Research Institute, AIST, Tsukuba 305-8568, Japan*

³*Department of Applied Chemistry,
University of Tokyo, Tokyo, 113-8656, Japan*

⁴*Physics Department, Brookhaven National Laboratory,
P. O. Box 5000, Upton, New York 11973, USA*

(Dated: February 2, 2008)

Abstract

A systematic analysis of the complex lineshape at $(\pi, 0)$ of heavily overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ is presented. We show that a coherent component in the quasi-particle excitation is correlated with the superfluid density throughout the entire doping range and suggest it is a direct measure of the order parameter of high temperature superconductors.

PACS numbers: 71.10.Ay, 74.72.Hs, 79.60.Bm

[‡] Present address: Department of Applied Physics, Stanford University, Stanford, California 94305

*Electronic address: dlfeng@fudan.edu.cn

The off-diagonal nature of the order parameter of a superconductor makes it difficult to be measured directly[1]. For conventional superconductors, the appearance of an energy gap in the single particle excitation spectrum characterizes the superconducting state; as a result, the observation of the gap is virtually equivalent to the observation of the superconducting order parameter. On the other hand, for high temperature superconductors (HTSC's), because of the pseudogap phenomenon in the normal state of underdoped systems, such a strict link between the gap and off-diagonal long range order (ODLRO) breaks. There have been suggestions between ODLRO and the (π, π) resonance peak in the dynamic structure factor measured by inelastic neutron scattering experiments around 40meV in HTSC's [2]. However, because a significant amount of spin fluctuations exist in this region and evolve with temperature in the normal state of underdoped systems, a direct and clean measurement of the superconducting order parameter is still not achieved.

We pointed out earlier that in the single particle excitation spectrum a sharp peak, which develops near $(\pi, 0)$ of the Brillouin zone below the superconducting transition temperature (T_c), exhibits similar behaviors as the superfluid density [3]. This so-called superconducting peak (SCP) is observed by angle-resolved photoemission spectroscopy (ARPES) in many HTSC's, first on $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212), later on $YBa_2Cu_3O_{7-\delta}$, and $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ [4]. Being a probe for single particle excitation, ARPES cannot measure collective properties such as the superfluid density (or phase stiffness) directly. However, it could be measuring condensate fraction, a single particle quantity closely related to superfluid density. Condensate fraction is defined as fraction of particles participating in pairing, and thus is the direct measurement of the ODLRO. Unlike the gap, the correlation between the SCP and superconducting order parameter works well in the underdoped regime. However, its validity in the overdoped regime has been questioned. Some claim that the SCP increases upon overdoping, while the superfluid density decreases[5]. Others claim that there is no SCP on the overdoped sample, and the peak-dip-hump (PDH) lineshape associated with the SCP is simply due to the bilayer band splitting (BBS) effect[6, 7, 8, 9].

In this paper, we present a global study on the photon energy ($h\nu$) and temperature (T) dependent ARPES spectra of heavily overdoped Bi2212 at $(\pi, 0)$. We show that there is a substantial SCP in the heavily overdoped regime and its correlation with the ODLRO holds across the entire doping range. This also gives a unified understanding of the PDH lineshape in the BBS circumstance[10]: the anti-bonding band (AB) and bonding band (BB)

each develop their own PDH lineshapes by transferring parts of their spectral weight into the newly-created coherent quasiparticles (AB peak and BB peak, respectively).

ARPES experiments were performed at beamline V-4 of Stanford Synchrotron Radiation Laboratory (SSRL). Data were collected with He-I light from a He discharge lamp (for the T -dependence) and polarized synchrotron light from a normal incidence monochromator (for the $h\nu$ -dependence), where the contamination to the spectra from the second order light is extremely weak, ideal for a quantitative comparison between spectra taken with different $h\nu$'s. Detailed descriptions of experimental settings and sample preparation can be found in Ref.[10].

The existence of the coherent peak has been discussed extensively for optimally doped and underdoped systems based on doping and T dependence[3]. Here we recap this with the data taken on an optimally doped Bi2212 ($T_c=90\text{K}$) sample in Fig. 1. As shown in Fig. 1a, the sharp coherent peak grows out of a smooth spectrum with decreasing temperature at $(\pi, 0)$. Fig. 1b shows how this peak behaves along $\Gamma - M$. We found that the SCP is strongest when the hump is within 120~150 meV below E_F ; away from this region, the SCP rapidly dies out while dispersing slowly with the hump to higher binding energies. The extracted superconducting state dispersions of the peak and hump in the main band are plotted in Fig. 1c. We emphasize that the SCP is not the AB, otherwise, one would obtain a bilayer splitting increasing towards $(0, 0)$ (Fig. 1d), while band theory as well as symmetry arguments gives zero splitting along $(0, 0) - (\pi, \pi)$ [11].

The complication in the heavily overdoped regime originates from the resolution of the AB and BB in the normal state, which forms a PDH-like lineshape with the sharp AB in the lowest binding energy position[10]. Kordyuk *et al.* have implemented a model fit of two bilayer-split bands to study the $h\nu$ -dependent superconducting state $(\pi, 0)$ spectra in this doping regime[7]. A qualitative agreement between the fitted $h\nu$ -dependent matrix elements of the peak and hump and those of bilayer-split bands predicted by theory[12, 13] was argued to justify the peak and hump being due to the AB and BB, at least in the overdoped regime[9]. However, we found that although the fitting function is able to fit each single spectrum well at different $h\nu$'s, it faces difficulty when more physical constraints are included. One example is shown in Fig. 2a. These spectra are taken at $(\pi, 0)$ on an overdoped $T_c=65\text{K}$ sample (OD65) at different temperatures. We start with the nine normal state EDC's to obtain the global parameters through a global fit (described below).

The fit gives an excellent agreement with the experiments which confirms the qualification of the chosen form of spectral function for describing the normal state features and, notably, the thermal sharpening-up process on the lineshapes upon T decrease. However, with these parameters, the discrepancy between the eight simulated and experimental superconducting state spectra increases upon going further into the superconducting state. This demonstrates a drastic lineshape variation beyond the description where only two bilayer-split bands are considered. The much more rapid intensity accumulation on the experimental peak feature below T_c requires further investigation.

Reflecting the behavior of the SCP in underdoped, optimally doped, and even slightly overdoped samples[14], we extend the fitting function in Ref.[7] by adding two spectral components to account for the rapid spectral weight accumulation near E_F .

$$I(\omega, T, h\nu) = I_0(T, h\nu) \cdot \left[\left(\sum_{\alpha}^4 J_{\alpha}(\omega, T, h\nu) \cdot f(\omega, T) \right) \otimes R(\omega, \Gamma'(h\nu)) + B(\omega, T) \right] + I_1(T, h\nu)$$

, where f is the Fermi function, R the $h\nu$ -dependent resolution Gaussian, B the momentum-independent but T -dependent empirical background function obtained by a separate fit on the featureless energy distribution curves (EDC's) around $(\pi/2, \pi/2)$ [15], I_0 and I_1 the linear intensity coefficients. The summation is over the spectral intensity of AB hump, BB hump, AB peak and BB peak, given by, respectively,

$$\begin{aligned} J_{ah}(\omega, T, h\nu) &= M_{ah}(h\nu) \cdot C_a(T) \cdot A_h(\omega, T, \alpha, \varepsilon_{ah}), \\ J_{bh}(\omega, T, h\nu) &= M_{bh}(h\nu) \cdot C_b(T) \cdot A_h(\omega, T, \alpha, \varepsilon_{bh}), \\ J_{ap}(\omega, T, h\nu) &= M_{ap}(h\nu) \cdot (1 - C_a(T)) \cdot A_p(\omega, \Gamma_a(T), \varepsilon_{ap}), \\ J_{bp}(\omega, T, h\nu) &= M_{bp}(h\nu) \cdot (1 - C_b(T)) \cdot A_p(\omega, \Gamma_b(T), \varepsilon_{bp}) \end{aligned}$$

, where M is the matrix element, C the remaining spectral weight in the hump, A_h and A_p the self-normalized spectral functions for the hump and the peak, respectively, given by

$$\begin{aligned} A_h(\omega, T, \alpha, \omega_0) &= \frac{\xi |\Sigma''(\omega, T)|}{(\omega - \sqrt{\omega_0^2 + \Delta_{sc}(T)^2})^2 + \Sigma''(\omega, T)^2}, \\ A_p(\omega, \Gamma(T), \omega_0) &= \frac{2\sqrt{\ln 2}}{\sqrt{\pi}\Gamma(T)} \exp[-(\frac{\omega - \omega_0}{\Gamma(T)/2})^2]. \end{aligned}$$

We adapt the empirical $\Sigma''(\omega, T) = \sqrt{(\alpha\omega)^2 + (\beta T)^2}$ from Ref.[7], where α and β are made globally fittable ($h\nu$ - and T -independent) to reflect the doping and impurity level of samples,

and ξ is a normalization factor. $\Gamma(T)$ is a T -dependent linewidth (FWHM). Note that three major extensions in the fitting function have been made. First, two spectral components for the SCP's are added with their Gaussian lineshapes justified by the STM results[16]; second, spectral weight transfer (SWT) ($= 1 - C_{a(b)}, 0 \leq C_{a(b)}(T) \leq 1$) is proposed for the formation of SCP, which is a pronounced character of many correlated systems and is also based on the empirical observation of the fact that the integrated spectral weight is roughly T -independent in the $(\pi, 0)$ region [17]; third, a realistic SCG opening extracted with the leading edge gap method on the same sample introduces an effective T -dependent energy shift to the bilayer-split bands in the superconducting state.

In order to retrieve reliable quantitative information from a multi-dimensional fit to a set of interconnected EDC's, we developed a non-linear least-squares fitting routine, which has been applied in the normal state fit in Fig.2a. The goal is to achieve a systematic understanding of both the $h\nu$ - and the T -dependent behavior of the lineshape. Physical constraints are strictly followed here, for example, in the $h\nu$ -dependent set, all functions of T are global variants to all EDC's at the same T while the functions of $h\nu$ (matrix elements) are locally specified, and in the T -dependent set, vice versa. The renormalized band energy position ε is always a global fitting parameter for the EDC's of samples at the same doping level. Without any manual tuning in the global parameters, the new fitting function can yield a robust, physically-constrained global fit for a large set of EDC's regardless of the initial values input.

It has been shown before that the intensity of the BB peak drops quickly when the BB disperses deep below the E_F at $(\pi, 0)$ [10]. For simplicity, we first neglect J_{bp} term, and achieve an excellent global fit for all the spectra shown in Fig. 2b-d simultaneously, which include the aforementioned T -dependence data and the $h\nu$ -dependence $(\pi, 0)$ data of a Pb-doped OD65 (Pb-OD65) taken above and below T_c . In Fig. 3a, the T -dependent energy positions of the maxima of three spectral features are shown. In the superconducting state, the AB peak is located quite close to the AB hump, which makes it very difficult to resolve them separately in experiments. For the AB (BB) hump maximum, the slight decrease (increase) above T_c is fully determined by the T -dependence of the self-energy with the fitted $\alpha = 0.715 \pm 0.006 eV^{-1}$ and $\beta = 2.57 \pm 0.03 eV^{-1}$ while the superconducting state is mainly affected by the opening of the gap. The spectral maximum shift in the BB hump is T -insensitive as compared with the AB hump; for the AB peak which turns up in the

superconducting state, its almost-constant location ($-18.01 \pm 0.03 \text{ meV}$) seems to define the maximum SCG opening at $(\pi, 0)$. This contradicts the scenario where the peak near E_F is just the AB which should change its position with the SCG opening. The extracted T -averaged BBS amplitude is $84.39 \pm 0.07 \text{ meV}$ [19], consistent with Ref.[10].

The fitted matrix elements as a function of $h\nu$ for the three spectral features are shown in Fig. 3b. Matrix elements of AB and BB hump vary in the same range of magnitude but with an anti-phase-like $h\nu$ -dependence as predicted by theory[12, 13]. Note that the self-normalization of the spectral function is required not only for a well-defined SWT analysis but also for a physically comparable matrix element between different spectral components. $\langle M_{bh} \rangle / \langle M_{ah} \rangle = 0.966 \pm 0.007$, while the matrix elements of AB peak shows a different $h\nu$ -dependence with those of the humps and ($\langle M_{ap} \rangle / \langle M_{ah} \rangle = 0.610 \pm 0.007$). This deviation is not unexpected because SCP is of redistributed spectral weight related to the superfluid, whose wavefunction may have different properties with the hump counterpart.

The T -dependent SWT from the above fit is summarized in Fig. 4a. The most prominent feature is the superconducting peak emerges only below T_c , while the SCP at lower doping could emerge 10K above T_c (see Fig. 1a and Ref.[3]). This may associate with the superconducting fluctuations in the lower doping regime[20, 21]. The SWT of AB is extrapolated to be 0.223 ± 0.028 at 0 K, a value far below 1, which casts doubt on the scenario where the entire lower-lying AB peak possesses almost all the weight of AB quasiparticle[22]. We also present the result of a 4-component fit with both J_{ap} and J_{bp} terms. The BB peak weight extrapolates at 0 K to be only 0.015 ± 0.008 , much smaller than that of AB, which also justifies the 3-component fit.

In Fig. 4a, the superfluid density measured by muon spin relaxation (μSR) at the same doping level is also displayed, which resembles the SWT in the T -dependence. In Ref.[3], we proposed a phenomenological subtraction of a smooth background to retrieve the SCP, which was further quantified by defining the so-called superconducting peak ratio (SPR) as the SCP intensity divided by the overall spectral weight in $(\pi, 0)$ spectrum taken at $h\nu=22.7 \text{ eV}$ and low temperature. Although the SWT is more physically well-defined than SPR in that it is free of the matrix element effects and takes into account the multi-component lineshape, when the hump is broad near or below optimal doping, SPR obtained by the phenomenological procedure reasonably characterizes the SCP. Thus, we calculated SPR for OD65 at $h\nu=22.7 \text{ eV}$ at 10K based on the globally fitted AB peak. It is plotted in Fig.

4b together with SPR's at lower doping levels reproduced from Ref.[3]. One can clearly see that SPR decreases with doping in the heavily overdoped regime, similar to the superfluid density[3, 23]. To account for this decrease, it has been suggested that the overdoped system is phase-separated into superconducting regions and hole rich normal metal regions[23]. In any case, these two intimate correlations between the SCP and superfluid suggest the SWT is a direct measurement of the superconducting order parameter.

We note that due to the lack of quantitative theories, the fitting formula used here is somewhat empirical, and certainly could be replaced by more sophisticated formulae with different number of components. However, the essence of SWT or any other equivalent is to capture the anomalous rapid spectral weight growth below T_c at the gap energy scale beyond the Fermi-liquid-based descriptions and to offer a direct measure of the superconducting order parameter which, in return, facilitates examination of various theoretical models. There have been a few theoretical proposals of such a quasiparticle peak created in the superconducting state by spectral weight transfer[24], and its weight was related to the condensate fraction or ODLRO. However, most of these theories are based on underdoped cuprates, while the overdoped regime is seldom addressed. The observed correlation between the SWT and superfluid density throughout the entire phase diagram calls for further theoretical efforts.

We thank Dr. D. H. Lu, and W. S. Lee at SSRL for experimental help, and K. M. Shen and Dr. A. Damacelli for comments and discussions. G.D.G. is supported by Department of Energy under contract No. DE-AC02-98CH10886. D.L.F. is supported by the National Science Foundation of China. SSRL is operated by the DOE Office of Basic Energy Science Divisions of Chemical Sciences and Material Sciences.

-
- [1] C. N. Yang, Rev. Mod. Phys. **34**, 694 (1962).
 - [2] S. Chakravarty and H.-Y. Kee, Phys. Rev. B **61**, 14821 (2000).
 - [3] D. L. Feng *et al.*, Science **289** 277 (2000).
 - [4] D. S. Dessau *et al.*, Phys. Rev. Lett. **66**, 2160 (1991); D. H. Lu *et al.*, *ibid.* **86**, 4370 (2001); D. L. Feng *et al.*, *ibid.* **88**, 107001 (2002).
 - [5] H. Ding *et al.*, Phys. Rev. Lett. **87**, 227001 (2001).

- [6] Z. M. Yusof *et al.*, Phys. Rev. Lett. **88**, 167006 (2002).
- [7] A. A. Kordyuk *et al.*, Phys. Rev. Lett. **89**, 077003 (2002).
- [8] S. V. Borisenko *et al.*, Phys. Rev. B **66**, 140509 (2002).
- [9] S. V. Borisenko *et al.*, Phys. Rev. Lett. **90**, 207001 (2003).
- [10] D. L. Feng *et al.*, Phys. Rev. Lett. **86**, 5550 (2001).
- [11] M. R. Norman *et al.*, Phys. Rev. B **52**, 615 (1995); A. I. Liechtenstein *et al.*, Phys. Rev. B **54**, 12505 (1996).
- [12] D. L. Feng *et al.*, Phys. Rev. B **65**, 220501(R) (2002).
- [13] M. Lindroos *et al.*, Phys. Rev. B **65**, 054514 (2002).
- [14] J. C. Campuzano *et al.*, Phys. Rev. Lett. **83**, 3709 (1999); A. Kaminski *et al.*, Phys. Rev. Lett. **86**, 1070 (2001).
- [15] We adopted a slightly different form of background approximation for a good match with the T-dependent EDC's. $B(\omega, T) = (1 + b_1\omega + b_2\omega^2 + b_3\omega^3) \cdot f(\omega - \sqrt{\Delta_b^2 + \Delta_{sc}(T)^2}, T + T_b)$, where $\Delta_{sc}(T)$ takes into account the superconducting gap (SCG) opening and the other parameters vary among experiments on different samples.
- [16] S. H. Pan *et al.*, Nature **413**, 282 (2001).
- [17] M. Randeria *et al.*, Phys. Rev. Lett. **74**, 4951 (1995).
- [18] Ch. Niedermayer *et al.*, Phys. Rev. Lett. **71**, 1764 (1993)
- [19] We define the BBS amplitude in the experimental sense, i.e., the energy separation between the maxima of AB and BB, which do not coincide with the renormalized band energy position ε .
- [20] Z. A. Xu *et al.*, Nature **406**, 486 (2000).
- [21] J. Corson *et al.*, Nature **398**, 221 (1999).
- [22] M. Eschrig and M. R. Norman, Phys. Rev. Lett **89**, 277005 (2002).
- [23] Y. J. Uemura *et al.*, Int. J. Mod. Phys. **B14**, 3703 (2000).
- [24] For example, see E. W. Carlson *et al.*, Phys. Rev. B **62**, 3422 (2000) for the stripe theory; D.-H. Lee, Phys. Rev. Lett. **84**, 2694 (2000) for the RVB gauge theory.

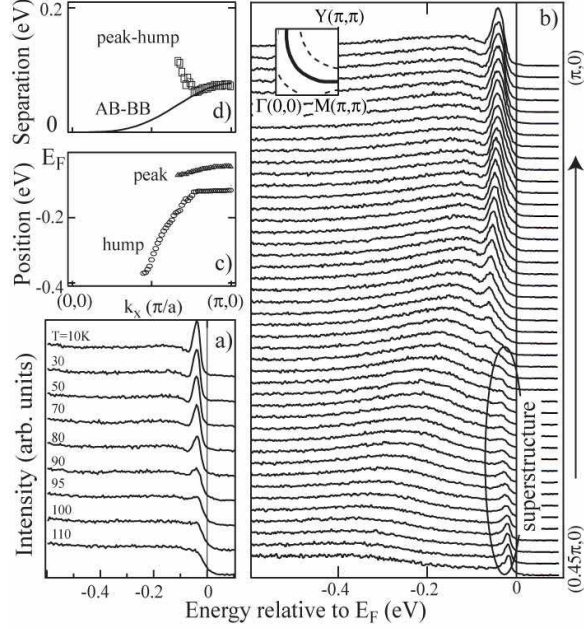


FIG. 1: ARPES spectra for optimally doped Bi2212 as a function of a) T and b) momentum in the superconducting state along $(0.45\pi, 0) - (\pi, 0)$. Features in the circled region are due to the superstructures. c) The extracted dispersions of the peak and hump based on the spectral maxima of features. d) The calculated peak-hump spacing is compared with the theoretical prediction for bilayer energy splitting in the superconducting state.

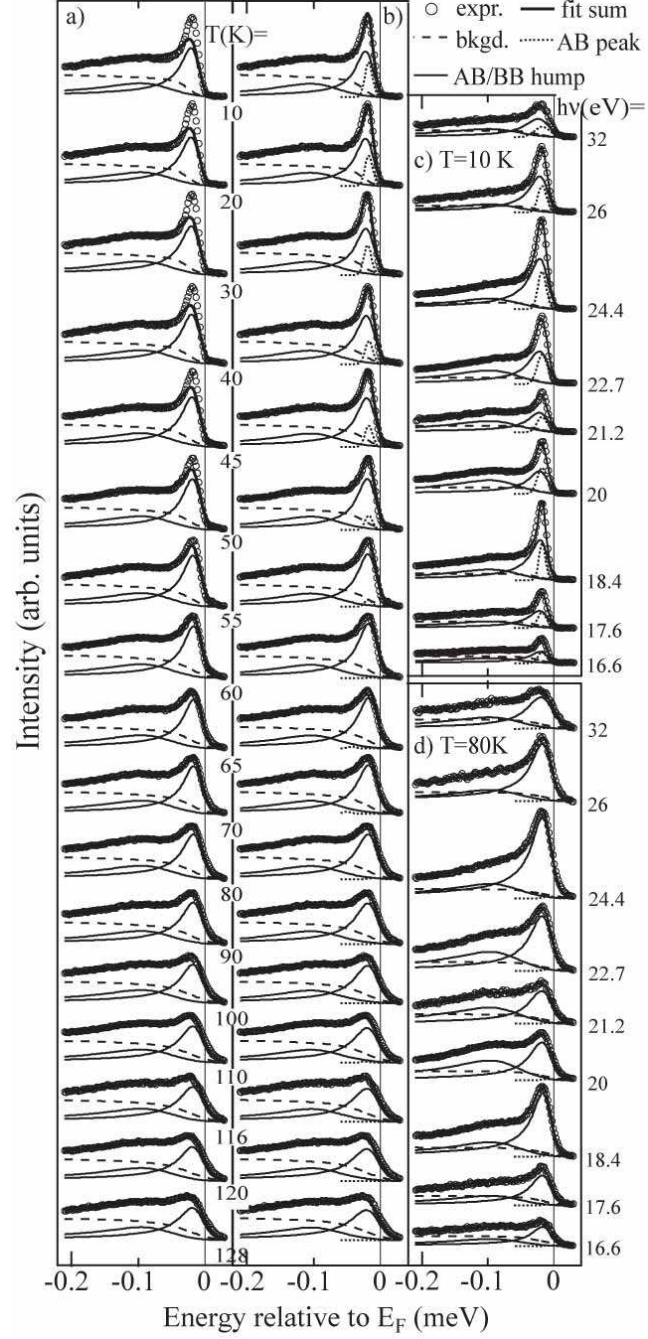


FIG. 2: T -dependent ARPES spectra at $(\pi, 0)$ of OD65 in (a) fitted by 2-component model, (b) by 3-component model including $h\nu$ -dependent spectra on Pb-OD65 at (c) $T=10$ K and (d) $T=80$ K (see text). The AB hump lies lower in energy than the BB hump. The width difference of the SCP is due to resolution variations from 10 to 18 meV at different $h\nu$'s.

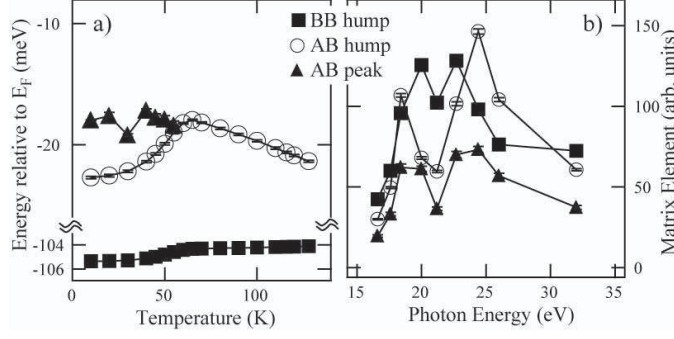


FIG. 3: The fitted a) T -dependent energy positions of the maxima of various features and (b) $h\nu$ -dependent matrix elements of different spectral features.

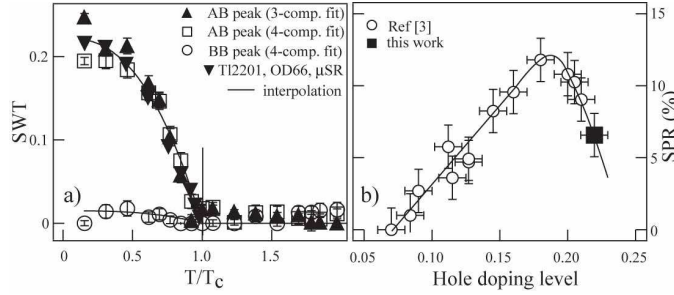


FIG. 4: (a) The T -dependent SWT's given by 3-component and 4-component fit are shown. The interpolations are based on a trial function $SWT = SWT_0[1 - (T/T_c)^\zeta]$ [18] on the T -dependence of SWT (averaged). The fitted value, $\zeta \sim 2.5$, agrees well with the T -dependent μ SR result on $Tl_2Ba_2CuO_{6+\delta}$ at the same doping level (OD66 Tl2201) (after Ref.[18]). (b) The doping-dependence of SPR at $h\nu=22.7$ eV supplemented by the global fit result on OD65.